

PATENT SPECIFICATION

(11) 1292718

1292718

NO DRAWINGS

(21) Application No. 5573/70 (22) Filed 5 Feb. 1970
 (31) Convention Application No. P 19 05 834.5
 (32) Filed 6 Feb. 1969 in
 (33) Germany (DT)
 (45) Complete Specification published 11 Oct. 1972
 (51) International Classification C05G 3/00; C07C 127/00
 (52) Index at acceptance

C1B 3C1 3C3 3C4 3C5 3CX 3F1 3G 4
 B2E 199 200 20X 20Y 30X 32X 32Y 33X 349 389 410
 417 433 439 43X 44Y 468 496 497 49Y 508
 532 599 620 627 643 649 64X 65Y 706 707 70Y
 718 742 802 803 804 80Y
 C1A C5A CSD28 D10 D28 G47 G47D28 G49 G49D10
 G50 G50D28
 C2C 20Y 303 30Y 341 34Y 578 626 799 KC



(54) A SALT OR FERTILIZER PROVIDED WITH A COATING
 TO PREVENT DUSTING OR CAKING

(71) We, BADISCHE-ANILIN- & SODA - FABRIK AKTIENGESELLSCHAFT, a German Joint Stock Company, of 5 6700 Ludwigshafen, Federal Republic of Germany, do hereby declare the invention, for which we pray that a Patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following Statement:—

10 Granular or crystalline salts and fertilizers can be coated with a powder to protect them against agglomeration. Suitable powders include finely divided lime, diatomaceous earth, talc, clays and the like and they are generally 15 used in amounts of up to about 2.5% by weight. Such powders do not however adhere well to the surface and dust is raised during movement in a warehouse, filling into bags and application of the fertilizer. Such dusts are a 20 nuisance, cause soiling and may even be a health hazard. The dust formation is especially troublesome during the filling of fertilizer or salt into bags made of plastics because these can then be heat-sealed only with difficulty if 25 at all. Adhesion of the powder to the fertilizer moreover cannot be substantially increased by adding water or steam or substances having adhesive properties such as mineral oils, glues, polysaccharide glycolic acid ethers (German 30 Patent No. 950,559) or condensation products of formaldehyde and urea or aminoplast-forming amines (German Patents Nos. 1,062,713 and 1,072,256), and therefore dust formation cannot be permanently prevented.

35 To avoid agglomeration, attempts have also been made to coat fertilizers and salts with mineral oils, paraffin waxes or mixtures of

these substances (French Patent No. 1,394,629). When these coating agents are used, the troublesome dust formation occurring in the case of powders is avoided but they have not been used in practise because of the inadequate anticaking effect when small amounts are used. The addition of large amounts of organic substances (more than 0.4%) in the case of fertilizers containing nitrates is ruled out because of the explosion hazard.

40 It has also been proposed to avoid the agglomerating properties of fertilizers by providing them with a coating of a water-insoluble polymerized material or treating them with a polymerizable material which can be converted (after it has been applied to the particles of fertilizer) into a polymerized material which is insoluble in water (British Patent No. 815,829). Examples of polymerized materials given are those which can be obtained by polymerization or copolymerization of styrene, vinyl chloride, vinylidene chloride, acrylonitrile, ethylene or fluorinated unsaturated hydrocarbons. The use of these coatings presupposes that the fertilizer is provided with a coating of an inorganic material, for example calcium sulfate, prior to the application of the said coating. The fertilizer may be first treated with calcium sulfate which is then cemented onto the surface of the fertilizer by adding water. Only then can the polymerized substances be applied, for example in the form of a solution in a suitable solvent. This method is fairly expensive because it requires at least two operations.

45 50 55 60 65 70 75
 It is also known from French Patent No.

1,394,629 that in order to prepare slow-release fertilizers they may be provided with a coating of a mixture of a paraffin wax having a low melting point and a polyalkylene having a molecular weight of not more than 10,000. The polyalkylene content may be from about 5 to 30% by weight and is generally not more than 50% by weight. These mixtures are usually applied to the fertilizer in an amount not exceeding 20% by weight, 10 to 18% by weight being used as a rule. Coatings of these mixtures penetrate fairly easily and deeply into the particles of fertilizer so that the release of nutrient is retarded in the desired manner.

For this reason and because of their plasticity such coatings have only a slight anticaking effect. In order to prevent caking of such fertilizers, particularly under tropical conditions, it is therefore necessary additionally to powder such fertilizers, for example with lime.

Finally there are numerous proposals known according to which the agglomeration of fertilizers and salts is to be prevented by adding surfactants. The proposals include sulfamic acid (German Patent No. 944,317), soaps, condensation products of formaldehyde with naphthalenesulfonic acid (Belgian Patent No. 529,255), alkylaryl sulfonates, aryl sulfonates, polyethylene glycol, amides, esters, sulfonamides, oleyl sarcosides, primary, secondary and tertiary alkylamines, alkylarylamines and arylamines and their salts.

The addition of these surfactants has only a limited effect however, particularly in the case of hygroscopic bulk goods such as fertilizers, because it cannot prevent the action of humidity and therefore it is necessary to powder the material as well in order reliably to prevent agglomeration.

According to another known proposal (German Printed Application No. 1,075,642), agglomeration of fertilizers is to be prevented by adding a cation-active amine or a salt thereof alone or in the form of a solution. Examples of suitable solvents are nitriles or mineral oils. When such solutions are used, agglomeration, particularly of hygroscopic fertilizers, under the influence of humidity cannot be prevented permanently.

The present invention seeks to provide coatings which improve the storage properties of granular or crystalline salts or fertilizers by reducing or preventing dust formation or agglomeration of the salts and fertilizers even under prolonged influence of humidity.

According to the invention the granular or crystalline salt or fertilizer is provided with a coating comprising a polyethylene wax and/or polypropylene wax having a penetrometer number (as defined herein) of not more than 3 and a mean molecular weight of from 500 to 10,000 and a surfactant, the amount of wax being from 0.01 to 1.5% by weight and the amount of surfactant being from 0.01 to 2% by weight of the total weight of the salt or fertilizer.

Salts and fertilizers which have been provided with a coating of a combination of one of the said waxes and a surfactant generally exhibit far better storage properties than those which have been provided with a coating of only one of the two components. These excellent storage properties cannot even be obtained by applying only one of the said components as a coating in a much greater amount than in the case of the combination according to the invention.

Polymers of ethylene or propylene, preferably having a mean molecular weight of from 3,000 to 10,000, are used as the waxes. The mean molecular weight is determined viscometrically according to K. Überrichter, "Die Makromolekulare Chemie", Vol. 8 (1952), pages 21-28. The waxes may also have undergone terminal oxidation. The melting point of these waxes is generally above 90°C. preferably from 100° to 110°C. The polyethylene wax, according to a preferred embodiment, has a degree of branching of 10 to 100 CH₂-groups per 1,000 carbon atoms. The penetrometer number of the wax is not more than 3, preferably 0 to 2, mm at 25°C and 100 g load after five seconds. The penetrometer number is determined according to test method DGF-M-III-96 of the Deutschen Gesellschaft für Fettmissenschaft, Münster, Westphalen, Germany.

In order to achieve particularly good storage properties in the fertilizers and salts the wax component is as a rule applied first to the solid to be coated. The wax may be applied in solid or liquid form, the solid to be coated advantageously having a temperature which is from 10°C below to 30°C above the melting point of the wax in question. The solid to be coated should preferably have a temperature which is up to 10°C above the melting point of the wax. In order to obtain a uniform coating which adheres well, the crystallized or granular solid is advantageously kept in motion and well mixed. For example by means of a rotating drum or a vibratory feeder.

In order to achieve a particularly good synergistic effect it is advantageous to limit the duration of the treatment at the abovementioned temperatures of application. Thus it is advantageous to carry out the treatment at the said temperatures within five to fifteen minutes. When using high temperatures, i.e. about 30°C above the melting point of the wax, about five minutes is chosen, while the treatment period can be extended to fifteen minutes at the lower limit of the said range of temperatures.

When the treatment is over, during which thorough mixing of the wax with the fertilizer or salt should be achieved, it is advantageous to cool the treated material rapidly, for ex-

ample at the rate of more than 3°C per minute, preferably more than 2°C per minute, to a temperature of about 40°C below the melting point of the wax in question. Cooling may 5 be carried out by a conventional method, for example in a cooling drum or fluidized bed. Following the treatment of the solid with the wax, the surfactant is added, if desired dissolved in a suitable organic solvent or emul- 10 sified in water, again with thorough mixing, the same equipment advantageously being used as for application of the wax. The application of the surfactant may be carried out prior to the cooling of the fertilizer or salt which 15 has been treated with wax, but is advantageously carried out after the said cooling. It is also possible to reverse the sequence by first applying the surfactant and only then treating the fertilizer or salt with wax. The 20 former method however usually gives better results. It is also possible to apply the wax and the surfactant to the solid simultaneously. If desired, it is possible to apply with the 25 surfactant a dye for identifying the solid, for example fertilizer, or to use a colored fertilizer or wax.

The surfactants may be any compounds which contain hydrophobic and hydrophilic 30 groups. A hydrophobic group usually consists of a long chain aliphatic hydrocarbon radical having at least four to thirty carbon atoms, advantageously eleven to twenty carbon atoms. The radical may however consist of an aryl 35 radical (for example a phenyl or naphthyl radical) or an alkyl-substituted aryl radical having up to thirty carbon atoms in the alkyl group. Apart from compounds having only one hydrophilic grouping in the molecule, compounds containing two or more such groups in the 40 molecule may also be used.

The hydrophilic groups may have an anionic or cationic nature. The carboxyl group ($-\text{COO}^-$), sulfo group ($-\text{SO}_3^-$, sulfonates) and the sulfate group ($-\text{OSO}_3^-$, sulfuric 45 esters) are particularly suitable anionic groups.

Instead of pure carboxylic acids, alkylsulfonic acids, arylsulfonic acids or alkylarylsulfonic acids or their salts and sulfuric esters, it is also possible to use those having substituents in the hydrophobic radical, for example with an amide, ester or sulfonamide group or ether oxygen. For example oleyl 50 sarcoside, the reaction product of oleyl chloride and the sodium salt of isethionic acid and a similar compound which is obtained from oleic acid and the sodium salt of N-methyltaurine may be used.

Fatty acids having eleven to twenty carbon 60 atoms and also aromatic acids such as especially benzoic acid or salts of these acids are particularly suitable as carboxylic acids. Purely aliphatic sulfonic acids having four to thirty, preferably eleven to twenty, carbon atoms, but also those having aromatic hydro- 65

phobic radicals, for example phenyl or naphthyl radicals, are also suitable as sulfonic acids. Naturally compounds having alkyl-substituted aromatic radicals are also possible. Alkylarylsulfonates which have been formed by condensation of naphthalenesulfonic acids with formaldehyde and which contain two or three naphthalene systems are particularly suitable. 70

Substances having cationic groups may also be used as well as those having anionic groups. Primary, secondary and tertiary amines and their salts or the corresponding quaternary ammonium salts are particularly suitable examples of these. The hydrophobic groups again consists of a hydrocarbon radical having at least 4 carbon atoms and up to twenty carbon atoms. It is advantageous to use amines however which contain at least eight carbon atoms. Not only are aliphatic amines such as octylamine, decylamine and stearylamine, cycloaliphatic amines such as dicyclohexylamine, cyclododecylamine and the like suitable, but also pyridinium salts, and not only morpholine, but also its N-alkyl or N-aryl derivatives having a total of six to twenty-one carbon atoms, for example the sulfate of N-phenylmorpholine. Aromatic amines, for example naphthylamine, are also suitable. Naturally amines having more than one amino group may be used, for example trimethylene-diamines having the general formula: 75

$$\text{R}-\text{NH}-\text{CH}_2-\text{CH}_2-\text{CH}_2-\text{NH}_2$$

where R denotes an aliphatic, cycloaliphatic or substituted aromatic hydrocarbon radical having six to twenty-two carbon atoms. 80

The last group of surfactants suitable for the coatings according to the invention are nonionic substances as obtained for example by the addition of ethylene oxide or propylene oxide to aliphatic alcohols or fatty acids having six to twenty carbon atoms, preferably more than eight carbon atoms, alkylphenols having at least one alkyl radical containing one to twenty carbon atoms, preferably eight to twenty carbon atoms. As a rule from 7 to 30, preferably from 7 to 20, moles of ethylene oxide or propylene oxide is required depending on the starting material. Examples are the oxyethylation product of 1 mole of octadecyl alcohol with 7 to 9 moles of ethylene oxide, or of 1 mole of p-nonylphenol with 9 to 12 moles of ethylene oxide. Polyethylene glycol having a molecular weight of more than 700 up to 8,000 is also suitable. 85

The said surfactants may be applied singly or mixed together to the salts and fertilizers. 90

The waxes are added to the crystallized or granular salts or fertilizers in amounts of 0.01 to 1.5% by weight, preferably 0.03 to 0.5% by weight, of the total weight of the salt or fertilizer. 95

All granular or crystallized substances cap- 100

105

110

115

120

125

able of being poured are suitable as salts and fertilizers. Application of the coatings according to the invention is particularly suitable to substances which have hygroscopic properties and therefore tend to agglomerate. Examples are: ammonium nitrate, mixed fertilizers containing ammonium nitrate such as calcium ammonium nitrate, calcium nitrate and multinutrient fertilizers, and also urea, potassium nitrate, nitrophosphates or multinutrient fertilizers containing urea, potassium nitrate, potassium sulfate, potassium chloride and sodium chloride.

The advantages of the coatings according to the invention are that they can be applied simply and rapidly to the substances to be

protected, usually in apparatus which is required in any case for the granulation. Even when used in small amounts, storage properties can be achieved with this combination of substances which cannot be achieved with far larger amounts of polyethylene waxes or surfactants known for the purpose in question. The additional use of powders is therefore unnecessary. Salts and fertilizers provided with the coatings according to the invention can be handled without dust formation and remain free-flowing even after prolonged storage.

The invention is illustrated by the Examples.

Examples

The fertilizers treated are as follows:

	Example	Fertilizer
35	1a, 1b, 1c, 1d	NPK Mg. fertilizer 12/12/17/2
	2	NPK fertilizer 15/15/15
	3	NPK fertilizer 12/12/20
	4a, 4b	NPK fertilizer 13/13/21
	5	NP fertilizer 20/20
40	6a, 6b	calcium ammonium nitrate
	7	NPK fertilizer 15/15/15
	8	NPK fertilizer 15/15/15
	9	calcium nitrate
	10	NPK fertilizer 13/13/21
45	11a, 11b	NPK fertilizer 13/13/21
	12a, 12b	NPK fertilizer 13/13/21
	13	potassium chloride
	14a, 14b	urea
	15	sodium nitrate
50	16	NPKMg fertilizer 16/11.5/14.5/2
	17	NPK fertilizer 10/8/10
	18	NPK fertilizer 14/9/21
	19	NPK fertilizer 15/9/14
	20	NPKMg fertilizer 10/15/20/2
	21	NPK fertilizer 15/15/15
55	22	NPK fertilizer 13/13/21
	23	potassium chloride
	24	NPK fertilizer 13/13/21
	25	NPKMg fertilizer 12/12/17/2
60	26	NPKMg fertilizer 12/12/17/2
	27	NPKMg fertilizer 12/12/17/2
	28	calcium ammonium nitrate
	29	NPK fertilizer 15/15/15
	30	NP fertilizer 20/20

The vertical columns in the following Table give the following data:

Ex=Example No.

Temp=temperature in °C

TT=treatment time in minutes

CAF=component added first (in % by weight)

CAS=component added second (in % by weight)

PT=pressure test described below (in kg)

BT=bag test described below

h=hard

lh=less hard

ff=free flowing

aff=almost free-flowing

mh=moderately hard

Individual samples are heated in a drum to the temperature (Temp), treated with the substance CAF and CAS, rolled for the time (TT) and cooled to about 60°C within fifteen minutes. After having been cooled, the samples prepared are subjected to the two tests described below:

1. Pressure test (PT)

A dismountable metallic cubic mold having an inside edge length of 70 mm and being open at the top is filled with the sample to be tested. The sample is then loaded for two hours at room temperature with a punch which transmits a force of 175 kg to the sample. The cubic press-molding formed is carefully removed from the mold and immediately broken

80

85

90

95

up by the action of pressure between two parallel plates in a destruction measuring instrument. The force indicated by the apparatus as being necessary for the destruction of the

5 press-molding is a measure of the tendency of the sample to agglomerate. High values denote a marked tendency to cake. In the ideal case, the press-molding breaks up when removed from the mold.

10 2. *Bag test (BT)*

In this test 50 kg of the material to be tested is stored in each case for six weeks in a bag which is impermeable to air and moisture and is loaded with ten times its weight. The bag is

15 then carefully opened and the material evaluated.

Under 1a and 1b in the Table below, coatings are specified with consist either of pure polyethylene wax or a surfactant. Under 1c

20 the results of the two tests are given which have been obtained with the combination of the two substance according to the invention for purpose of comparison. With a total amount of only 0.2% by weight with reference

25 to the multinutrient fertilizer, results are ob-

tained which as may be seen from the Table cannot nearly be achieved with five times the amount of the individual components.

The following further abbreviations are used in the following Table:

30

PEW 1a=polyethylene wax having a molecular weight of 5000, a degree of branching of 30 to 50 CH₂ groups per 1000 carbon atoms;

35

PEW=polyethylene wax

(s), (l) or (m)=applied in the form of a solid, liquid or melt

CP=a condensation product of 2 moles of β -naphthalenesulfonic acid with 1 mole of formaldehyde;

40

MW=molecular weight;

CP 10/1=condensation product of 10 moles of ethylene oxide and 1 mole of p-nonylphenol;

45

CP 8/1=condensation product of 8 moles of ethylene oxide and 1 mole of octadecyl alcohol;

PPW=polypropylene wax;

PEG=polyethylene glycol;

TABLE

Ex	Temp	TT	CAF	CAS	PT	BT
1a	108	10	none	none	520	h
	108	10	0.05 PEW 1a(s)	none	150	1h
	108	10	0.10 PEW 1a(s)	none	100	1h
	108	10	0.50 PEW 1a(s)	none	130	1h
	108	10	1.0 PEW 1a(s)	none	200	1h
1b	108	10	none	none	520	h
	108	10	0.05 CP	none	210	h
	108	10	0.10 CP	none	80	1h
	108	10	0.50 CP	none	90	1h
	108	10	1.00 CP	none	80	1h
1c	108	10	0.1 PEW 1a(s)	0.10 CP	10	ff
1d	108	20	0.10 PEW 1a(s)	0.10 CP	50	ff
2	112	10	0.10 CP	0.12 PEW 1a(s)	5	ff
3	100	10	0.10 CP	0.15 PEW 1a(m)	3	ff
4a	108	10	0.15 PEW 1a(s)	0.10 2-ethylhexylamine	10	aff
4b	108	10	0.25 2-ethylhexylamine	none	90	1h
5	105	10	0.08 PEW 1a(s)	0.02 stearylamine	2	ff
6a	110	10	0.10 PEW 1a(l)	0.30 dicyclohexylamine	3	ff
6b	110	10	0.40 dicyclohexylamine	none	100	mh
7	108	10	0.10 PEW 1a(l)	0.10 N-phenylmorpholine	8	aff
8	108	10	0.20 PEW 1a(l)	0.10 n-butylamine	12	aff
9	97	10	1.50 PEW MW 3 500	1.00 di-2-ethylhexylamine	3	ff

Ex	Temp	TT	CAF	CAS	PT	BT
10	108	10	0.2 PEW 1a(l)	0.15 cyclododecylamine	4	ff
11a	110	10	0.10 PEW 1a(l)	0.20 calcium stearate	15	aff
11b	110	10	0.50 calcium stearate	none	120	mh
12a	105	10	0.15 PEW 1a(s)	0.10 CP 10/1	20	aff
12b	105	10	0.10 CP 10/1	none	150	h
13	130	5	0.10 stearic acid	0.20 PPW MW6000	2	ff
14a	100	15	0.10 benzene sulfonic acid	0.10 PEW 1a	10	ff
14b	100	15	0.10 PEW MW7000	none	50	mh
15	105	15	0.10 PEW 1a	0.05 PEG MW3000	5	ff
16	110	10	0.1 dodecyl sulfonate	0.12 PEW MW4000(s)	8	ff
17	108	15	0.12 PEW MW550(s)	0.05 sodium benzyl-naphthalene sulfonate	10	aff
18	108	10	0.15 PEW MW4000(s)	0.08 sodium diisobutyl-naphthalene sulfonate	7	ff
19	113	5	0.07 stearylamine	0.10 PEW MW7000(l)	9	ff
20	112	10	0.12 PEW MW3000(l)	0.08 lauric acid	5	ff
21	109	15	0.05 Na-salt of mono-palmitic ester of sulfuric acid	0.15 PEW MW5000(l)	8	ff
22	107	15	0.08 PEW MW1900(s)	0.10 morpholine	10	aff
23	100	12	2.0 caproic acid	0.05 PPW MW3400(l)	10	ff
24	115	10	0.5 CP 8/1	0.05 PEW MW9000(s)	5	ff
25	110	10	0.1 PPW MW3300(l)	9.08 melissic acid	15	aff
26	118	10	0.1 calcium arachidate	0.1 PPW MW9000(s)	10	ff
27	112	12	0.1 N-ethyl-morpholine	1.0 PPW MW5000(s)	8	ff
28	110	10	0.2 PEW MW7000(l)	0.15 N-stearyl-morpholine	10	ff
29	110	8	0.5 PEW MW6000(s)	0.08 sodium octodecane-1-sulfonate	12	ff
30	102	8	0.15 benzoic acid	0.1 PPW MW8000(l)	0	ff

WHAT WE CLAIM IS:—

5 1. A granular or crystalline salt or fertilizer provided with a coating for the avoidance of dust formation and agglomeration wherein the coating comprises a polyethylene wax and/or polypropylene wax having a mean molecular weight of from 500 to 10,000 and a penetrometer number (as hereinbefore defined) of not more than 3, and a surfactant, the amount of

wax being from 0.01 to 1.5% by weight and the amount of surfactant being from 0.01 to 2% by weight of the total weight of the salt or fertilizer.

2. A salt or fertilizer as claimed in claim 1 wherein the polyethylene wax used has a degree of branching of from 10 to 100 CH₃ groups per 100 carbon atoms.

3. A salt or fertilizer as claimed in claim 1

or 2 wherein the surfactant is an anionic substance.

4. A salt or fertilizer as claimed in claim 3 wherein the anionic substance consists of a water-repellent hydrocarbon radical and an anionic group, the hydrocarbon radical being an aliphatic hydrocarbon radical having four to thirty carbon atoms, a phenyl radical, a naphthyl radical, an alkyl-substituted phenyl radical or an alkyl-substituted naphthyl radical and the anionic group being a carboxyl, sulfato or sulfonic group.

5. A salt or fertilizer as claimed in claim 1 or 2 wherein the surfactant is a cationic substance.

6. A salt or fertilizer as claimed in claim 5 wherein the cationic substance is a primary, secondary or tertiary amine or a salt thereof or a quaternary ammonium salt having an aliphatic radical containing four to twenty carbon atoms or a naphthyl radical.

7. A salt or fertilizer as claimed in claim 5 wherein the cationic substance is morpholine or a N-alkyl or N-aryl derivative of morpholine having a total of six to twenty-one carbon atoms.

8. A salt or fertilizer as claimed in claim 1 or 2 wherein the surfactant is a reaction product of ethylene oxide with an aliphatic alcohol having 6 to 20 carbon atoms and/or a fatty acid having 6 to 20 carbon atoms and/or an alkyl phenol, having at least one alkyl radical containing 1 to 20 carbon atoms.

9. A salt or fertilizer as claimed in claim 1 or 2 wherein the surfactant is a polyethylene glycol having a molecular weight of from 700 to 8000.

10. A salt or fertilizer as claimed in claim 1 and described in any of the foregoing Examples.

11. A process for the production of a granular or crystalline salt or fertilizer provided with a coating for the avoidance of dust formation and agglomeration as claimed in any of claims 1 to 10 wherein a polyethylene wax and/or polypropylene wax having a mean molecular weight of from 500 to 10,000 and a penetrometer number as hereinbefore defined) of not more than 3 is applied to the granular or crystalline salt or fertilizer whose temperature lies within a range from 10°C below to 30°C above the melting point of the wax to be applied and after mixing is cooled to a temperature which is 40°C below the melting point of the wax and then the surfactant is applied.

12. A process as claimed in claim 11 carried out substantially as described in the foregoing Examples.

J. Y. & G. W. JOHNSON,

Furnival House,

14—18, High Holborn, London, W.C.1,

Chartered Patent Agents,

Agents for the Applicants.

Printed for Her Majesty's Stationery Office by the Courier Press, Leamington Spa, 1972.
Published by the Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from
which copies may be obtained.

THIS PAGE BLANK (USPTO)